Efficient transport of fossil organic carbon to the ocean by steep mountain rivers: An orogenic carbon sequestration mechanism

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ABSTRACT

Mountain building exposes fossil organic carbon (OC_{fossil}) in exhumed sedimentary rocks. Oxidation of this material releases carbon dioxide from long-term geological storage to the atmosphere. OC_{fossil} is mobilised on hillslopes by mass wasting and transferred to the particulate load of rivers. In large fluvial systems it is thought to be oxidised in transit, but in short, steep rivers that drain mountain islands, OC_{fossil} may escape oxidation and re-enter geological storage due to rapid fluvial transfer to the ocean. In these settings, the rates of OC_{fossil} transfer and their controls remain poorly constrained. Here we quantify the erosion of OC_{fossil} from the Taiwan mountain belt, combining discharge statistics with measurements of particulate organic carbon load and source in 11 rivers. Annual OC_{fossil} yields in Taiwan vary from 12 ± 1–

246 ± 22 tC km^{-2} yr^{-1}, controlled by the high physical erosion rates that accompany rapid crustal shortening and frequent typhoon impact. Efficient transfer of this material ensures that $1.3 \pm 0.1x10^6$ tC yr^{-1} of OC_{fossil} exhumed in Taiwan is delivered to the ocean, with <15% loss due to weathering in transit. Our findings suggest that erosion of coastal mountain ranges can force efficient transfer and long-term re-accumulation of OC_{fossil} in marine sediments, further enhancing the role of mountain building in the long-term storage of carbon in the lithosphere.

INTRODUCTION

About $15x10^{15}$ tC of carbon is stored in rocks as fossil organic matter. This is almost 400 times the amount of carbon present in the atmosphere and oceans (Sundquist and Visser, 2004). The balance between the growth of this geological reservoir through burial of newly photosynthesised organic matter, and its decrease through oxidation of OC_{fossil} plays a crucial role in the long-term evolution of atmospheric CO_2 and O_2, and thus global climate (Berner, 1982; Berner and Canfield, 1989; Derry and France-Lanord, 1996; Hayes et al., 1999). It is commonly assumed that during mountain building, exhumed OC_{fossil} is completely converted to CO_2 by chemical weathering (Lasaga and Ohmoto, 2002; Bolton et al., 2006). OC_{fossil} can escape oxidation when physical erosion delivers it to the solid load of mountain rivers (Kao and Liu, 1996; Blair et al., 2003; Leithold et al., 2006; Hilton et al., 2008a). But when it enters large river systems (>100,000 km^2 area), up to 85% is oxidised in transport (Galy et al., 2008a; Bouchez et al., 2010). In contrast, short mountain rivers that drain to the ocean could deliver OC_{fossil} more efficiently to marine basins due to rapid transport in turbid waters (Dadson et al., 2005; Hilton et al., 2008b). Despite its potential importance, the transfer of OC_{fossil} from mountain islands has remained poorly constrained (Blair et al., 2003), due to both a lack of constraint of the source of particulate organic carbon (POC) in river sediments in these settings (Stallard, 1998; Lyons et al., 2002) and its transport behavior over the large range of water discharges in steep mountain catchments (Blair et al., 2003; Hilton et al., 2008a). To address this issue, we have determined the source of POC and the relation between OC_{fossil} transport and water discharge in rivers draining the mountain belt of Taiwan.

STUDY AREA, SAMPLING AND METHODS

Located along the western edge of the Pacific Ocean, mountain building in Taiwan is driven by collision between the Luzon Arc on the Philippine Sea plate and the Asian continental margin since 7 Ma (Teng, 1990). Steep rivers draining Taiwan's Central Range pass over narrow coastal plains to the ocean (Dadson et al., 2003). Inside the mountain belt, they have incised Mesozoic and Cenozoic siliciclastic and carbonate rocks, which have been metamorphosed up to greenschist and amphibolite facies (Ho, 1986). These rocks contain on average 0.2% OC_{fossil}, mainly of marine origin (Hilton et al., 2010). The western flank of the Central Range comprises Late Cenozoic turbiditic mudstones, sandstones and near-shore foreland sediments (Ho, 1986). These lithologies contain on average 0.4% mainly terrestrial OC_{fossil} (Hilton et al., 2010). Metamorphic grade decreases from East to West across the Central Range and surface rocks contain OC_{fossil} of varying thermal maturity and structure, ranging from poorly organized carbonaceous matter to polycrystalline graphite (Beyssac et al., 2007). Due to rapid crustal shortening (Teng, 1990) and the prevailing subtropical cyclonic climate, rates of mass wasting and physical erosion in river catchments of the Central Range are exceptionally high, averaging ~6 mm yr^{-1} over the last four decades (Dadson et al., 2003), and Taiwan Rivers supply 384×10^6 t yr^{-1} of suspended sediments and ~120×10^6 t yr^{-1} of river bed load to the ocean.

To determine the concomitant OC_{fossil} transfer, we measured suspended sediment concentration (SSC, mg L^{-1}), OC_{fossil} concentration in the particulate load (POC_{fossil}, mg L^{-1}) and water discharge (Q_w, m^3 s^{-1}) in 11 main Taiwan Rivers over an 18 month period following established methods (Dadson et al., 2003; Hilton et al., 2008b; Hilton et al., 2010). The catchments ranged in size from 310 km^2 to 2,906 km^2 (covering a total area of 9.6×10^3 km^2, 27% of the islands surface) and were sampled 1–3 times per month over two typhoon seasons (March 2005-September 2006) to cover the dynamic range of Q_w. All catchments drain more than one of the major geological formations, sourcing rocks with OC_{fossil} content ranging between 0.2% and 0.4% (Hilton et al., 2010). Determination of the source of POC in each sample has been described in detail elsewhere (Hilton et al., 2010). Briefly, an end-member mixing model was used to quantify the fraction of OC_{fossil} (F_f) in the total POC using measurements of the nitrogen to organic carbon ratio and the stable carbon isotopes of organic matter. F_f was tested against independent constraints from radiocarbon, and F_f average precision and accuracy are 0.09 and 0.05, respectively. POC_{fossil} for a suspended sediment sample is the product of SSC, total organic carbon concentration and F_f.

To quantify river solid load yields we defined rating curves that link the Q_w measured at a station to the river load constituent concentration (SSC and POC_{fossil}, mg L^{-1}), and applied them to the continuous daily record of Q_w at that station to estimate the mass transfer of suspended load materials over the sampling period. Following common practice for small catchments, we used power law rating curves (Fig. 1a) with a least squares best fit to available data (Hilton et al., 2008b). Quoted errors on mass transfer estimates combine the rating curve exponent error (Fig. 1b) and the error in F_f (Hilton et al., 2010).

FLUVIAL TRANSPORT OF OC_{fossil} AND ITS CHEMICAL ALTERATION

In all rivers, OC_{fossil} was present in the suspended load throughout the sampling period. General, positive relationships between measured Q_w, and SSC and POC_{fossil} in these rivers are described well by power laws (Fig. 1a) with very similar least squares best fit exponents for SSC and POC_{fossil} in a given catchment (Fig. 1b). The link between OC_{fossil} and suspended sediment confirms their common rock source in mountain catchments (Leithold et al., 2006; Hilton et al., 2008a).

Following the observed relationship between POC_{fossil} and Q_w and the derived power law rating curves, OC_{fossil} yields for all rivers ranged between 12 ± 1 and 246 ± 22 tC km^{-2} yr^{-1} over the gauged period (Fig. 2). These yields are significant natural transfers of carbon and two rivers had OC_{fossil} yields >225 tC km^{-2} yr^{-1}, greater than the highest total POC yield (fossil+non-fossil) previously reported for mountain rivers (Stallard, 1998; Lyons et al., 2002; Hilton et al., 2008a). The average OC_{fossil} yield for the 11 studied catchments was 82 tC km^{-2} yr^{-1}.

High OC_{fossil} yields in Taiwan are closely linked to the yield of suspended sediment (Fig. 2) and are therefore controlled by physical erosion rate. This concurs with previous findings which suggest OC_{fossil} is delivered to river channels by mass wasting, e.g., bedrock landslides (Hilton et al., 2008a), and gully erosion (Leithold et al., 2006), processes which can drive rapid physical erosion rates in mountain belts. Here, erosion of OC_{fossil} must occur faster than its chemical alteration (Petsch et al., 2000), leading to incomplete oxidation in the weathering zone. The lack of substantial sediment storage in the bedrock channels of the Central Range implies that the fluvial transit time is very short (Dadson et al., 2005), further restricting alteration of OC_{fossil} during transport, in notable contrast to large river systems (Galy et al., 2008a; Bouchez et al., 2010). OC_{fossil} weathering may still occur within catchments, since sediment may reside for longer periods of time in regoliths and soils on hillslopes where the production of mineral surface area through physical erosion may enhance OC_{fossil} weathering rates (Petsch et al., 2000; Bolton et al., 2006; Lasaga and Ohmoto, 2002).

OC_{fossil} oxidation in catchments can be quantified by comparing the measured fluvial export of OC_{fossil} to that predicted by eroding surface bedrock at known erosion rates. Measured fluvial exports imply a OC_{fossil} content of 0.35 ± 0.03% (±σ) in suspended sediments from Taiwan (Fig. 2). Surface rocks have an average organic carbon content of 0.24 ± 0.19% (±σ, *n* = 31) which varies between geological formations (Hilton et al., 2010). The highest average OC_{fossil} content of the main geological formations is 0.41%, which is similar to the +σ bound of all samples. If we assume little variability of OC_{fossil} content with grain size, demonstrated by previous work (Galy et al., 2008a; Bouchez et al., 2010; Hilton et al., 2010), this can be used to estimate that a maximum of 15 ± 7% of the exhumed OC_{fossil} is weathered prior to export (Fig. 2). This moderate weathering loss would correspond to a transfer of geological carbon to the modern hydrosphere and atmosphere of 12 ± 6 tC km^{-2} yr^{-1} in the sampled catchments. However, the measured average OC_{fossil} in rock precludes any weathering loss and is within one standard deviation of the OC_{fossil} content of suspended sediments (Fig. 2) making it difficult to estimate chemical alteration of OC_{fossil} by this method. We conclude that the bulk of exhumed OC_{fossil} is exported to the ocean from Taiwan in river sediment and that the magnitude of OC_{fossil} oxidation requires further investigation.

DELIVERY OF OC_{fossil} TO THE OCEAN

We estimate that the sampled mountain rivers delivered 0.9×10^6 tC yr^{-1} of OC_{fossil} to the ocean during the study period (Fig. 3). To quantify the OC_{fossil} transfer from the island over a longer period, we note that mean sediment yields in sampled catchments were 23,600 ± 6,800 t km^{-2} yr^{-1} (± standard error on the mean) during our study, and 21,700 ± 3,900 t km^{-2} yr^{-1} in the period 1970-1999 (Dadson et al., 2003) suggesting the measured OC_{fossil} yields are a natural feature of this mountain belt. On decadal timescales the spatial pattern of physical erosion in

Taiwan is set by the incidence of earthquakes and typhoons, and by bedrock erodability (Dadson et al., 2003), and this is likely also the case for the erosion of OC_{fossil}. We therefore combine the published decadal suspended sediment transfer (Dadson et al., 2003) with the average OC_{fossil} concentration in the suspended load (0.35 ± 0.03% calculated by regression across 11 catchments; Fig. 2) to estimate a total fluvial export of $1.3 \pm 0.1 \times 10^6$ tC yr^{-1} of OC_{fossil} to the ocean as suspended sediment. This equates to a normalized yield of 37 ± 3 tC km^{-2} yr^{-1} over the total area of Taiwan (35,980 km^2). Addition of bed load transport, assuming an average OC_{fossil} content of 0.27 ± 0.12% (±σ) measured in 14 river catchments (Hilton et al., 2010), results in a total export of ~1.7×10^6 tC yr^{-1}. The suspended flux alone represents ~1% of the estimated 90-240x10^6 tC yr^{-1} total POC (fossil+non-fossil) input to the oceans (Stallard, 1998; Lyons et al., 2002) from only ~0.02% of Earth's landmass.

OC_{fossil} transported through the modern erosion system can be re-buried in long-lived marine sediments (Dickens et al., 2004; Galy et al., 2008a). Locally this can alter the geochemical record of the organic matter because OC_{fossil} has a variable isotopic signature (Hayes et al., 1999; Hilton et al., 2010). If OC_{fossil} re-burial is globally significant then it will influence the residence time of carbon in the lithosphere and our understanding of the long-term cycles of carbon and oxygen (Berner and Canfield, 1989; Galy et al., 2008a), while also influencing our interpretation of the isotopic mass balance of carbon in the oceans (Derry and France-Lanord, 1996; Hayes et al., 1999). In large river systems with long transport pathways and significant sediment storage, only refractory graphitic OC_{fossil} is resilient to chemical weathering and physical attrition during transport (Galy et al., 2008a). In the case of the Madeira floodplain of the Amazon, >85% of the eroded OC_{fossil} may escape geological storage to the atmosphere (Bouchez et al., 2010). In contrast, the steep mountain rivers of Taiwan export

OC_{fossil} eroded from rocks with a range in thermal maturity (Hilton et al., 2010), with little OC_{fossil}-loss across the island irrespective of its graphitization state (Fig. 2).

The fate of OC_{fossil} exported from Taiwan is not well constrained, but several observations suggest that a significant proportion is re-buried in marine sediments. First, by its nature OC_{fossil} is associated with mineral surfaces, and this has been shown to enhance organic carbon burial efficiency (Hedges and Keil, 1995). Second, offshore Taiwan rapid accumulation of clastic sediment is likely to optimize organic carbon preservation (Canfield, 1994). Hyperpycnal sediment discharge in very turbid river plumes, and deposition of turbidites may be especially important in this process (Hilton et al., 2008b). Hyperpycnal discharge represented 30%–42% of the sediment export from Taiwan to the ocean in 1970–1999, and may be even more important on longer time scales (Dadson et al., 2005). Accounting for bed load transport, and assuming full preservation of hyperpyncal OC_{fossil} on long time scales, this results in a re-burial flux of 0.5–$0.7x10^{6}$ tC yr^{-1} in basins around Taiwan. The closest constraint on the fate of OC_{fossil} comes from the Images core MD012403 collected from the Okinawa Trough to the NE of Taiwan (Fig. 3). There, the radiocarbon age of bulk organic carbon in sediments is offset by ~7,000 years from the age of planktonic foraminifera, throughout the Holocene (Kao et al., 2008). Assuming a binary mixture of radiocarbon-dead OC_{fossil} and contemporaneous organic matter, the authors estimate that of the ~0.7% organic carbon in sediments at this site, ~0.3% is OC_{fossil}. If this material is present even at a site ~150km from Taiwan's coastline and with limited hyperpycnal input, then our re-burial estimate of 0.5–$0.7x10^{6}$ tC yr^{-1} is likely to be conservative.

WIDER IMPLICATIONS AND CONCLUSIONS

To assess their wider significance, our findings can be compared to observations from a much larger orogenic system. Erosion of the Himalaya is thought to have resulted in re-burial of 0.3–0.5x10^6 tC yr^{-1} of OC_{fossil} in the deep marine Bengal fan (Galy et al., 2008a). Averaged over the source area, the OC_{fossil} re-burial flux in the Bengal fan represents 0.2–0.3 tC km^{-2} yr^{-1}. The equivalent re-burial flux estimated here for Taiwan is 14–19 tC km^{-2} yr^{-1} which is likely to represent a lower bound as discussed previously. This large discrepancy is due in part to a lower OC_{fossil} content in Himalayan surface rocks, typically <0.20% (Galy et al., 2008a), associated with older, higher-grade Proterozoic to Early Paleozoic meta-sediments. The discrepancy is also related to erosion rates which are 2–3 times higher in Taiwan (Galy and France-Lanord, 2001; Dadson et al., 2003). However, these factors cannot explain the factor ~70 difference in the normalized OC_{fossil} re-burial flux. Its main cause is the transit length and time of OC_{fossil} in the terrestrial environment. While fluvial entrainment and delivery of sediment to the ocean typically occur within a single flood event in steep rivers of Taiwan (Dadson et al., 2005; Hilton et al., 2008b), Himalayan sediment is routed through the Gangetic plain, with a large capacity for sediment storage and subsequent OC_{fossil} alteration (Galy et al., 2008b), as such only the most refractory components of OC_{fossil} persist at the river mouth (Galy et al., 2008a).

Our data suggest that where sedimentary bedrock is prevalent and clastic sediment yields exceed 3,000 t km^{-2} yr^{-1} (Fig. 2), OC_{fossil} should be present in river sediments. These conditions are met throughout the mountainous islands of Oceania, and on active margins throughout the world (Milliman and Syvitsky, 1992). If our findings from Taiwan apply more widely to those settings, then one effect of mountain building on the organic carbon cycle may be felt through the repeated exhumation, erosion and re-burial of previously sequestered CO_2 and the inhibition of its reflux to the atmosphere. This effect is likely to be governed disproportionately by re-burial of OC_{fossil} in basins adjacent to steep, coastal mountain ranges. At present, the combined OC_{fossil} re-burial flux in the Taiwanese and Himalayan source-to-sink systems is at least $0.8–1.2\text{x}10^{6}$ tC yr^{-1}, accounting for >1% of the present day total organic carbon burial in marine sediments (Berner, 1982; Schlünz and Schneider, 2000). Globally, this flux is presently unaccounted for in models of carbon cycling and atmospheric evolution (Berner and Canfield, 1989; Derry and France-Lanord, 1996; Lasaga and Ohmoto, 2002; Bolton et al., 2006;), yet should be sustained during orogenesis and contribute to geological storage of carbon derived from the atmosphere.

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FIGURES CAPTIONS

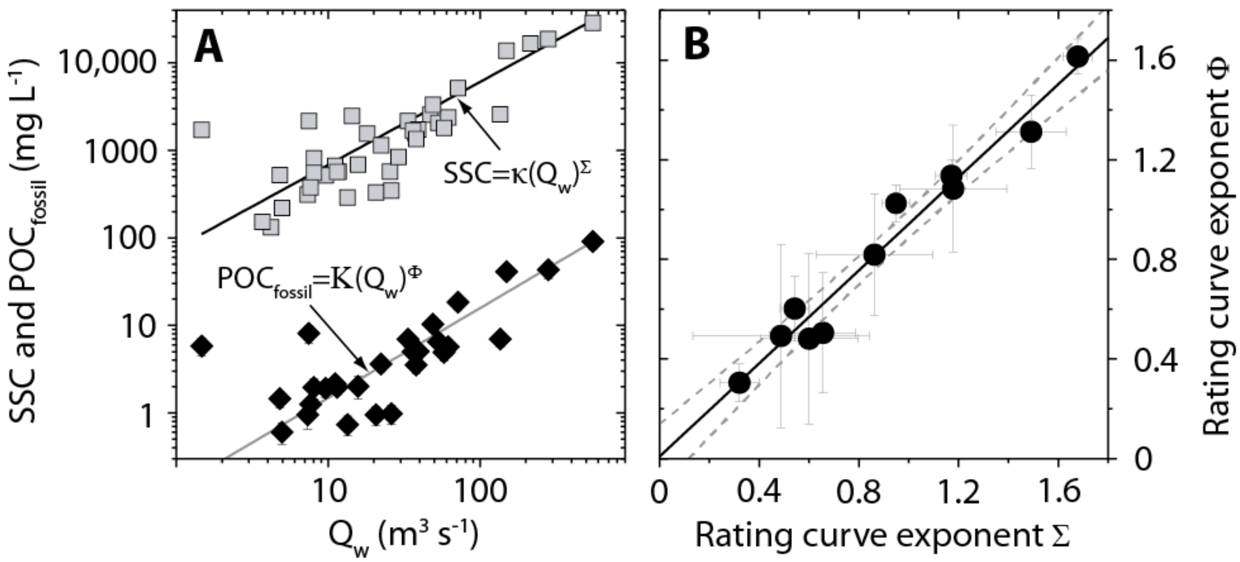

Figure 1. Relationships between water discharge (Q_w, m^3 s^{-1}) and fossil particulate organic carbon concentration (POC_{fossil}, mg L^{-1}) and suspended sediment concentration (SSC, mg L^{-1}) in Taiwan Rivers. (a) Direct measurements of Q_w, SSC and POC_{fossil} for the Chenyoulan River in Taiwan. Whiskers show error in concentration where large than the point. Power law rating curves for SSC (black line) and POC_{fossil} (gray line) were determined by a least squares best fit with exponents Σ and Φ, respectively. (b) Power law rating curve exponent between Q_w and solid load constituents (Σ and Φ) for 11 Taiwanese rivers determined by a least squares best fit. Whiskers are errors on the fit. Solid line show linear regression through the data (y = (0.93 ± 0.06)x + 0.01 ± 0.06; R^2 = 0.97, P<0.0001) and dashed lines 95% confidence intervals.

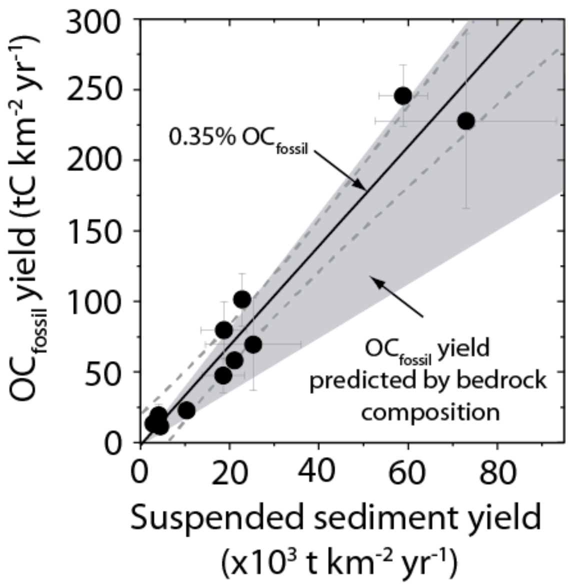

Figure 2. Relationship between suspended sediment yield (t km^{-2} yr^{-1}) and fossil organic carbon (OC_{fossil}) erosion yield (tC km^{-2} yr^{-1}) in Taiwan Rivers. A linear regression of the data (y = $(0.0035 \pm 0.0003)x - 1 \pm 10$; $R^2 = 0.94$ $P<0.0001$), dashed gray showing 95% confidence intervals, implies an average OC_{fossil} concentration of 0.35 ± 0.03% in suspended sediments and that clastic sediment transfer is the dominant control on OC_{fossil} yield. Shaded region (gray) indicates the predicted range of OC_{fossil} yields for the measured suspended sediment yields using the OC_{fossil} concentration measured in rock samples from the major geological formations in Taiwan (Hilton et al., 2010).

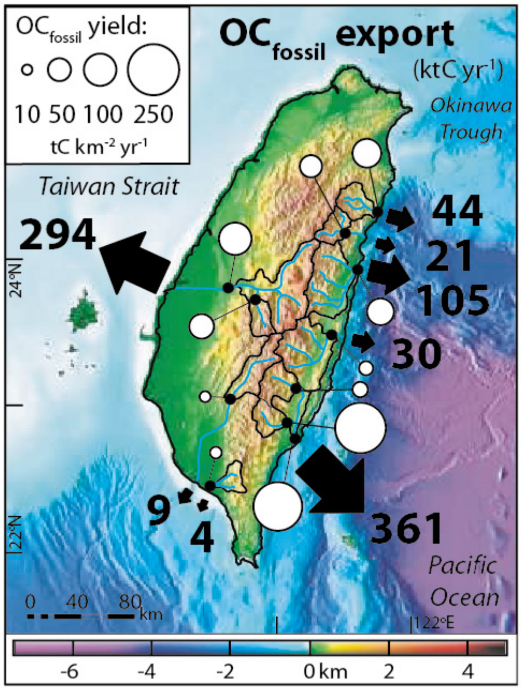

Figure 3. Fossil organic carbon (OC_{fossil}) export (ktC yr^{-1}) to the ocean from Taiwan over the sampling period. The sampling locations and gauging stations (black circles), their catchment area (black line) and main rivers (blue line) are overlain on topography bathymetry. The relative magnitude of the OC_{fossil} yield is indicated by the circles (gray).